Detailed *L* Emission Spectra and Satellites of $_{74}W$

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The $L\alpha$ and $L\beta$ spectra of tungsten were measured using a high resolution single crystal x-ray spectrometerand were fitted into Lorentzians. The fit residuals of the $L\alpha$ and $L\beta$ spectra indicate satellites in the vicinity of each spectrum, which are originated from Coster-Kronig transitions. Linewidths, energies, and intensities were estimated for each diagram line.

Keywords : $L\alpha,\beta$ satellite lines/ Coster-Kronig transition / spectator hole / natural linewidth / multiplet fitting/

It is generally difficult to analyse *L* x-ray emission spectra induced by electron impact because all the three *L* subshells can be ionized and the redistribution of initial vacancies due to Coster-Kronig transitions produce additional inner-shell vacancies. The existence of two or more holes in atomic inner shells gives rise to satellite lines with energies which are shifted from the diagram lines. The study of L x-ray satellites in high-Z elements has not been performed by many workers. There have been reported only a few papers on tungsten $_{74}W$ [1,2,3] for the last 20 years. The widths of some *L* x-ray lines of W were measured as part of a program for compiling the *L* series linewidths in heavy elements [4,5,6]. They suggested that the disagreement between theory and experiment without the analysis of hidden satellites was due to the large values of *M*- and *N*- subshells partial widths reported in the work of McGuire [7].

In the present study, we investigated the line width, energies and intensities including satellites of W $L\alpha,\beta$ emission lines generated by electron bombardment using a single-crystal high resolution x-ray spectrometer for the evaluation on the correctness of the theoretical calculation of different types of transition rates.

The tungsten *L* spectral lines were excited by electron bombardment in a rotating anode at tube voltage of 49 kV and 150-180 mA. The spectral measurements were carried out with a single-crystal spectrometer with symmetrical Si(440), Si(444) and Ge (444) perfect crystals in which x-ray topography showed no dislocation. A double slit collimator of 100 mm length and the vertical width of 10 or 20 mm was used for the measurement.

The natural linewidths (Γ_n) of the emission spectra were evaluated from the measured spectral linewidths (Γ_o) including the energy dispersion due to the slit (δE_{slit}) and the crystal ($\delta E_{crystal}$), as follows:

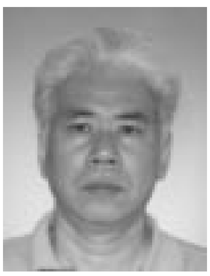
$$\Gamma_{nat}^{(n)} = \Gamma_{obs}^{(n)} - (\delta E_{slit} + \delta E_{crystal})^{(n)}$$

where (n) is the correction parameter, which was determined for each spectral line by numerically solving the above equations using two different experimental conditions of collimator slit or Bragg reflection. The obtained correction parameter (n) values between 1.4 < n < 1.6 were considered as reasonable in our experiment.

STATES AND STRUCTURE — Atomic and Molecular Physics —

Scope of Research

In order to obtain fundamental information on the property and structure of materials, the the electronic states of atoms and molecules are investigated in detail using X-ray, SR, ion beam from accelerator and nuclear radiation from radioisotopes. Theoretical analysis of the electronic states and development of new radiation detectors are also performed.



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The natural width of an x-ray line originated from a transition from an atomic level (A) to a level (B) is represented as the sum of the width of the initial and final levels: $\Gamma(A\rightarrow B) = \Gamma(A) + \Gamma(B)$

Theoretical values of natural width of atomic vacancy states were reported for *L*-shell [8], and *M*-shell [8] using the relativistic calculations, and for *N*-shell with nonrelativistic calculations [9].

The relative intensities of Coster-Kronig satellite transitions are calculated assuming that the radiative transition rate $\omega_{i=1,2,3}$ has the same value for single vacancy states and multiple vacancy states. For the transitions to the initial vacancy state L_2, the relative intensities of the spectator hole satellite lines to the diagram line can be calculated as follows:

$$Is(L_2)=(\sigma_1/\sigma_2)\, f_{1,2}\, P(L_1L_2X)$$

where $\sigma_{i=1,2,3}$ is the ionization cross section by electrons for the L_i subshell; $f_{i,j}$ is the partial Coster-Kronig transition probability from L_i to L_j level; $P(L_iL_jX)$ is the probability of the radiationless transition $L_i\rightarrow L_jX$, for which the double vacancy state L_jX is created, where X is either *M* or *N* shell. In the transitions to the initial vacancy state L_3, there are two possible Coster-Kronig transitions: $L_1\rightarrow L_3X$ and $L_2\rightarrow L_3X$. The relative intensities of the spectator hole satellite lines to the diagram line has two terms:

$$Is(L_3) = (\sigma_1/\sigma_3)\, f_{1,3}\, P(L_1L_3X) + (\sigma_2/\sigma_3)\, f_{2,3}\, P(L_2L_3X).$$

We used for ω_i the values reported by Krause [10], for σ_i the values reported by Reusch [11] for $_{73}$Ta at 50 kV, and for $P(L_iL_jX)$ the values reported by Chen *et al.* [12].

In Table 1 we compare our results with the previous experimental values by Salem *et al.*[4,5]. The values derived from theoretical linewidth as the sum of the two levels involved in the transition are shown for a comparison with the experimental values obtained with the multiplet fitting method by Deutsch *et al.* [13] by considering the satellite structure: the relative intensities and the relative energy position of the satellite lines to the diagram lines were fixed in the fitting. The energies of the satellites were taken from the table by Parente [14].

In the transitions involving *L*-shell levels and *M*-shell levels, such as $L\alpha_1$, $L\alpha_2$, $L\beta_1$, $L\beta_3$, and $L\beta_4$, we have a good agreement between the values obtained by multiplet fitting and the theoretical values which are obtained using the relativistic calculations. On the other hand, for the transitions involving *L*-shell levels and *N*-shell levels, such as $L\beta_2$, $L\beta_{15}$, $L\gamma_1$, the widths obtained from theoretical calculations are all larger than those from the experiments, the difference being between 2.7 eV and 3.3 eV. This disagreement between theory and experiment for $L_i\rightarrow N_j$ transitions can be attributed to the values for the *N* shell vacancy states obtained from nonrelativistic calculations. Although Gokhale *et al.* [6] and Salem & Lee [4] also suggested

this, they did not considered the satellite structure for each spectra.

The relative intensities derived from the radiative transition probabilities published by Scofield [15] are shown in Table 1 together with the experimental values reported by Salem *et.al* and our experimental values obtained by single Lorentzian fitting.

The structure left in the residue of fitting by using single Lorentzians may be attributed to the presence of the hidden satellites. Even after considering the satellite structure, by using the multiplet fitting, there still remains some structure in the residue. This situation has appeared for all analysed spectra, excluding $L\beta_{2,15}$, when the residue was completely solved. This can be due to the fact that we could not include in the multiplet fitting model the contribution of the: *O* and *P* shell spectator hole, and the shake-off processes.

References

[1] L.Salgueiro, M.L.Carvalho, and F.Parente, *Journal de Physique*, colloque C9, **48**, 609 (1987)

[2] A.N.Nigam, R.B.Mathur *Chemical Physiscs Letters* **28**, 41 (1974)

[3] A.N.Nigam, R.B.Mathur, and R.Jain, *J.Phys.***B7**, 2489

[4] S.I.Salem, and P.L.Lee, *Phys.Rev*.**A10**, 2033 (1974)

[5] S.I.Salem, S.L.Panossian, and R.A.Krause, *At. Data Nucl. Data Tables* **14**, 91 (1974)

[6] B.G.Gokhale *et. al*, *Phys.Rev.***A28**, 858 (1983)

[7] E.J.McGuire, *Phys.Rev.***A5**, 1043 (1972); **A6,** 851 (1972); **A10** 13 (1974)

[8] M.S.Chen, B.Crasemann, and H.Mark, *Phys.Rev.***A24**, 177 (1981); **A27**, 2989 (1983); **A21**, 449 (1980)

[9] E.J.McGuire, *Phys.Rev.***A9**, 1840 (1974)

[10] M.O.Krause, *J.Phys.Chem.Ref.Data* **8**, 307 (1979)

[11] S.Reusch, H.Genz, W.Low, A.Richter, *Z.Phys.***D3**, no.4, 379 (1986)

[12] M.H.Chen, B.Crasemann, H.Mark, M.Aoyagi, and H.Mark, *At.Data and Nuclear Data Tables*, **24**, 13 (1979)

[13] M.Deutsch, G.H"olzer, J.H"artwig, J.Wolf, M.Fritsch, and E.F"orster, *Phys.Rev.***A51**, 283 (1995)

[14] F.Parente, *At.Data Nucl.Data Tables* **26**, 383 (1981)

[15] J.H.Scofield, *At. Data Nucl. Data Tables* **14**, 121 (1974)

Table 1

Line	Transition	Linewidth							Relative Intensity		
		Experimental			Theoretical				Theoretical	Experiment	
		Salem [eV]	Present work	(n corr.) [eV]	Γi [eV]	Γf [eV]	Γi+Γf	Multiplet fitting	Scofield	Salem	Present work
WLα_1	L3-M5	7.84	7.022	(1.33)	4.812	1.795	6.607	6.675	100	100	100
WLα_2	L3-M4	5.27	7.27	(2.25)	4.812	1.868	6.680	6.784	11.34	11.16	11.43
WLβ_2	L3-N5	9.26	10.572	(1.603)	4.812	7.720	12.53	9.616	17.74	-	20.79
WLβ_{15}	L3-N4	-	7.73	(1.65*)	4.812	7.89	12.70	9.43	1.98	-	1.55
WL $\beta_{2,15}$									19.64	22.74	22.34
WLβ_6	L3-N1	-	11.65	-	4.812	15.00	19.81	-	1.246	1.25	1.25
WLβ_1	L2-M4	7.82	6.941	(1.54)	4.821	1.868	6.689	6.620	100	100	100
WLγ_1	L2-N4	10.20	10.626	(1.96)	4.821	7.89	12.71	10.07	18.61	18.80	23.16
WLβ_3	L1-M3	12.6	12.72	(1.52)	5.959	10.560	16.519	-	100	100	100
WLβ_4	L1-M2	13.20	14.66	(1.22)	5.959	11.90	17.859	-	79.88	67.8	77.32